(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 294 770 A2**

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 88109075.7

(22) Date of filing: 07.06.88

(51) Int. Cl.4: C08L 23/12 , C08L 23/08 , //(C08L23/12,23:08)

(30) Priority: 08.06.87 US 59053

(43) Date of publication of application: 14.12.88 Bulletin 88/50

(84) Designated Contracting States: AT BE CH DE ES FR GB GR IT LI LU NL SE

(71) Applicant: UNION CARBIDE CORPORATION
39 Old Ridgebury Road
Danbury Connecticut 06817(US)

(72) Inventor: Rifi, Mahmoud Rashad
28 Dawson Road
Kendall Park New Jersey 08824(US)

(74) Representative: Weinhold, Peter, Dr. et al
Patentanwälte Dr. V. Schmied-Kowarzik
Dipl.-Ing. G. Dannenberg Dr. P. Weinhold Dr.
D. Gudel Dipl.-Ing. S. Schubert Dr. P. Barz
Siegfriedstrasse 8
D-8000 München 40(DE)

(54) Impact polypropylene.

(57) An impact polypropylene composition comprising:

(i) isotactic polypropylene; and

(ii) a copolymer of ethylene and an alpha-olefin comonomer having 3 to 8 carbon atoms, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 20 to about 35 percent by weight based on the weight of the copolymer and (b) being present in the composition in the range of about 20 to about 50 percent by weight based on the weight of the polypropylene.

EP 0 294 770 A2

IMPACT POLYPROPYLENE

Technical Field

This invention relates to an improved version of impact polypropylene.

Background Art

Impact polypropylene is widely used in such applications as appliances, automobiles, furniture, and luggage. It is generally a blend of isotactic polypropylene with ethylene/propylene or ethylene/propylene/diene rubbers or a similar combination prepared in situ. The rubber modifier overcomes the low temperature brittleness and the lack of impact resistance of the isotactic polypropylene. However, when the polypropylene is modified with an amorphous rubber, it experiences a severe stress whitening or blush on impact, e.g., when an object made of this impact polypropylene is struck, a white discoloration appears. This is obviously undesirable as it limits the applications of the impact polypropylene.

Disclosure of the Invention

An object of this invention, therefore, is to provide a polypropylene, which exhibits much reduced blushing together with high impact resistance.

Other objects and advantages will become apparent hereinafter.

According to the present invention, the above object is met by a composition comprising the following components:

(i) isotactic polypropylene; and

(ii) a copolymer of ethylene and an alpha-olefin comonomer having 3 to 8 carbon atoms, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 10 to about 30 percent by weight based on the weight of the copolymer and (b) being present in the composition in a range of about 20 to about 50 percent by weight based on the weight of the polypropylene.

Detailed Description

Isotactic polypropylene homopolymer can be prepared by the process described in United States patent 4,304,891, issued on December 8, 1981, which is incorporated by reference herein. The homopolymer preferably has a melt flow in the range of about 1 to about 20 and xylene solubles in the range of about 2 to about 6.

The ethylene/alpha-olefin copolymer is the result of the copolymerization of ethylene and an alpha-olefin comonomer having 3 to 8 carbon atoms. It can be prepared by the processes described in European Patent Application 0 120 501 and 0 120 503, both published on October 3, 1984 and incorporated by reference herein. The density of the copolymer is in the range of about 0.87 to about 0.90 gram per cubic centimeter. The portion of the copolymer attributed to the alpha-olefin comonomer is in the range of up to about 40 mole percent based on the total number of moles in the copolymer and is preferably in the range of about 7 to about 30 mole percent. The balance of the copolymer is based on ethylene. The preferred comonomers are propylene, 1-butene, 1-hexene, and 1-octene. The portion of the copolymer based on comonomer is in the range of about 15 to about 60 percent by weight based on the weight of the copolymer, and is preferably in the range of about 20 to about 45 percent by weight. These ethylene/alpha-olefin copolymers are considered to be soft polymers because they are semi-crystalline. The crystallinity is in the range of about 20 to about 35 percent by weight. The copolymer has a melt index in the range of about 0.1 to about 10 grams per 10 minutes and preferably in the range of about 0.3 to about 1.0 gram per 10 minutes. Melt index is determined by ASTM D-1238, Condition E. It is measured at 190 °C. The isotactic polypropylene and the ethylene/alpha-olefin copolymers are, of course, not reactive with one another.

Subject composition can be prepared either by physical blending or in situ incorporation of the copolymer in a polypropylene matrix. The proportion of ethylene/alpha-olefin copolymer can be in the range

of about 20 percent by weight to about 50 percent by weight based on the weight of the isotactic polypropylene.

A typical blending technique is described as follows: isotactic polypropylene is mixed (dry) with the ethylene/propylene copolymer and the dry mixture is extruded at about 200° C and pelleted. Conventional stabiliz rs for the isotactic polypropylene and the ethylene/propylene copolymers are used.

A typical in-situ method is carried out in the following manner: isotactic polypropylene homopolymer is prepared in one reactor and is transferred to another reactor (or reactors) where the ethylene/propylene copolymer is produced in the presence of the polypropylene homopolymer. The in-situ mixture is taken out of the reactor(s), stabilized, and extruded at about 200° C and pelleted.

Various conventional additives can be added in conventional amounts to subject compositions such as antioxidants, ultraviolet absorbers, antistatic agents, pigments, dyes, fillers including carbon black, slip agents, fire retardants, stabilizers and smoke inhibitors.

The invention is illustrated by the following examples:

Examples 1 to 6

A polypropylene homopolymer having a melt flow of 4.0 and xylene solubles of 3.5 is blended with an ethylene/propylene copolymer or an ethylene/1-butene copolymer wherein the portion of the copolymer based on comonomer, propylene or 1-butene, is present in amounts of 25 and 15 percent by weight, respectively, based on the weight of the copolymer. The copolymer is present in the blend in an amount of 15 to 50 percent by weight based on the weight of the polypropylene. The blend is extruded and pelletized with the following stabilizers: 0.125 weight percent calcium stearate and 0.125 weight percent antioxidant. The pellets are injection molded into 125 mil specimens for evaluation.

The Table sets forth the variables, i.e., the weight percent of the copolymer based on the weight of the composition; the density of the copolymer in gram per cubic centimeter; and the crystallinity in percent by weight, and the results.

The tests used to obtain the results are as follows:

1. The Gardner Impact (-30° C) test is carried out according to ASTM D-3029. This test involves dropping a weight from a defined height onto an injection molded disk, 125 mil thick, which has been preconditioned at -30° C. The results are given in inch-pounds (inch-lbs).

2. The 1% SFM (Secant Flexural Modulus) test is carried out according to ASTM D-790. The results are given in pounds per square inch (psi).

3. The blushing (10 lbs) test is carried out as follows. An injection molded disk is used. The apparatus is the same as for the Gardner Impact test except that the falling weight (a steel bar) is 10 pounds. The steel bar is dropped on the disk. The disk is then aged at room temperature for 24 hours. Stress-whitening or blushing appears on the disk in the form a circle. The extent of stress-whitening is defined by the diameter of the circle measured in fractions of an inch (in).

4. Crystallinity is measured by Differential Scanning Colorimeter (DSC) using a Dupont 990 analyzer with a pressure DSC cell.

Table

Examples	Comonomer	% Co-Polymer	Density (g/cc)	Crystallinity (% by wt.)	Gardner Impact (inch-lbs)	1% SFM (psi)	Blushing (10 lb)(in)
1	propylene	30	0.86	<2	>320	110,000	0.63
2	propylene	15	0.87	20	20	160,000	0.5
3	1-butene	30	0.87	20	>300	125,000	0.4
4	1-butene	50	0.87	20	>320	86,000	0.0
5	1-butene	30	0.90	35	175	120,000	0.26
6	1-butene	30	0.92	45	20	130,000	<0.1
7.	1-butene	30	0.96	89	<10	186,000	0.0

Melt flow is determined in accordance with ASTM-1638.

Xylene solubles are defined as the fraction that stays in solution after the polypropylene sample is dissolved in hot xylene and the solution is allowed to cool to 23° C.

Claims

1. An impact polypropylene composition comprising:
(i) isotactic polypropylene; and
(ii) a copolymer of ethylene and an alpha-olefin comonomer having 3 to 8 carbon atoms, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 20 to about 35 percent by weight based on the weight of the copolymer and (b) being present in the composition in the range of about 20 to about 50 percent by weight based on the weight of the polypropylene.

2. The composition defined in claim 1 wherein the molded product thereof exhibits a blush of at most about 0.5 inch per 10 pounds.

3. The composition defined in one or both of the claims 1 to 2 wherein the portion of the copolymer based on comonomer is present in the range of about 15 to about 60 percent by weight based on the weight of the copolymer.

4. The composition defined in claim 3 wherein the portion of the copolymer based on comonomer is present in the range of about 20 to about 45 percent by weight based on the weight of the copolymer.

5. The composition defined in one or more of the claims 1 to 4 wherein the comonomer is 1-butene.

6. The composition defined in one or more of the claims 1 to 4 wherein the comonomer is propylene.

(19) Europäis h s Patentamt
European Patent Office
Office eur péen des brevets

(11) Publication number: 0 294 770 A3

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 88109075.7

(22) Date of filing: 07.06.88

(51) Int. Cl.4: C 08 L 23/12
C 08 L 23/08
//(C08L23/12,23:08)

(30) Priority: 08.06.87 US 59053

(43) Date of publication of application:
14.12.88 Bulletin 88/50

(84) Designated Contracting States:
AT BE CH DE ES FR GB GR IT LI LU NL SE

(88) Date of deferred publication of search report:
22.03.89 Bulletin 89/12

(71) Applicant: UNION CARBIDE CORPORATION
39 Old Ridgebury Road
Danbury Connecticut 06817 (US)

(72) Inventor: Rifi, Mahmoud Rashad
28 Dawson Road
Kendall Park New Jersey 08824 (US)

(74) Representative: Weinhold, Peter, Dr. et al
Patentanwälte Dr. V. Schmied-Kowarzik Dipl.-Ing. G. Dannenberg Dr. P. Weinhold Dr. D. Gudel Dipl.-Ing. S. Schubert Dr. P. Barz Siegfriedstrasse 8
D-8000 München 40 (DE)

(54) Impact polypropylene.

(57) An impact polypropylene composition comprising:
(i) isotactic polypropylene; and
(ii) a copolymer of ethylene and an alpha-olefin comonomer having 3 to 8 carbon atoms, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 20 to about 35 percent by weight based on the weight of the copolymer and (b) being present in the composition in the range of about 20 to about 50 percent by weight based on the weight of the polypropylene.

EP 0 294 770 A3

Bundesdruckerei Berlin

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 88 10 9075

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
X	GB-A-1 346 234 (MITSUI PETROCHEMICAL) * Claims 1-4; page 1, lines 72-85 *	1-6	C 08 L 23/12 C 08 L 23/08 // (C 08 L 23/12 C 08 L 23:08)
A	GB-A-2 060 658 (ASAHI-DOW) * Abstract *	1	
A	EP-A-0 052 557 (BP CHIMIE SOCIETE ANONYME TOUR NEPTUNE - LA DEFENSE) * Abstract *	1	
A	EP-A-0 168 129 (TOA NENRYO KOGYO K.K.) * Abstract *	1	
A	GB-A-2 063 278 (ASAHI-DOW) * Abstract *	1	

TECHNICAL FIELDS SEARCHED (Int. Cl.4)

C 08 L

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	28-12-1988	GOOVAERTS R.E.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P0401)